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A Time-Dependent Ginzburg-Landau Phase Field Formalism for Shock-Induced Phase Transitions

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Abstract. Phase-field models have become popular in the last two decades to describe a host of free-boundary problems. The strength of the method relies on implicitly describing the dynamics of surfaces and interfaces by a continuous scalar field that enters the global grand free energy functional of the system. Here we explore the potential utility of this method in order to describe shock-induced phase transitions. To this end we make use of the Multiphase Field Theory (MFT) to account for the existence of multiple phases during the transition, and we couple MFT to a hydrodynamic model in the context of a new LLNL code for phase transitions, SAMSA. As a demonstration of this approach, we apply our code to the $\alpha - \epsilon$ -Fe phase transition under shock wave loading conditions and compare our results with experiments of Jensen *et. al.* [*J. Appl. Phys.*, 105:103502 (2009)] and Barker and Hollenbach [*J. Appl. Phys.*, 45:4872 (1974)].

INTRODUCTION

In this short article we describe a time dependent Ginzburg-Landau [1] method, akin to what is also called the phase field method [2, 3] for specific situations, where tracking the dynamics of interfaces among different coexisting phases is of interest. Historically, the phase-field method is seen as a by-product of coarse grained models that describe large-scale behavior of matter near critical points for mainly second-order phase transitions, classified as models A,B and C in the scheme of Hohenberg and Halperin [4]. These non-equilibrium models use a local Gibbs free-energy that is dependent on different thermodynamic fields as well as an order parameter field $\phi(r, t)$. The latter demarcates the boundary (interface) between different phases, and through its time-dependence the description of the dynamics of the phase transitions is possible. In Figure 1 we depict a microscopic presentation of the order parameter ϕ for the case of solidification.

The kinetic equations for the field parameter ϕ are called non-conserved when their average value does not need to be satisfied by a conservation equation (unlike mass, momentum and energy). These are categorized as model A. An example is the magnetization at each finite volume element in an Ising magnet or the density of superconducting phase in a type-II superconductor [1](density of Cooper pairs at each finite volume). The total free-energy of the system can be written in the following expansion

$$F = \int dV \left[\frac{\epsilon}{2} |\nabla \phi|^2 + a\phi^2 + b\phi^4 \right], \quad (1)$$

where $\phi(r, t)$ is the density of Cooper pairs.

Kinetic equations which govern the evolution of ϕ are called conserved, and correspond to model B, if they take the form of a flux conserving equation. This is for example the case of the dynamics of the total solute concentration in a closed system. Model C will categorize non-equilibrium systems where the dynamics of non-conserved fields are coupled with the dynamics of one or more conserved fields. A classic example for which the phase-field model was first introduced is the case of the solidification of an undercooled and/or supersaturated liquid alloy. In the case of the solidification, the system will be described by the dynamic of the order parameter phase field (nonconserved) coupled with the dynamic of latent heat (conserved) generated at the solid-liquid interface as well as the dynamic of concentration (also conserved) [2].

Our ultimate aim is to explore the application of such methods in the realm of phase transitions initiated by dynamic compression. One of first detailed descriptions of the time-dependence of a dynamically-driven phase transition was the pioneering work of Boettger and Wallace on the shock-induced $\alpha - \epsilon$ transition in iron [5]. Here, the time derivative of the phase fraction was assumed to be related to the distance from thermodynamic equilibrium; this model has the advantage of containing essentially two independent parameters – a barrier height that governs metastability, and a relaxation timescale. For ramp compression experiments of iron, where the temperature can be significantly lower than in a shock wave, Bastea *et. al.* [6] undertook classical nucleation theory (CNT) analysis of the $\alpha - \epsilon$ Fe transition measured under quasi-isentropic loading on the Sandia Z machine. More recently, Yu *et. al.* [7] developed a phase-field model, akin to the aforementioned C-model in the Hohenberg-Halperin scheme, but coupled its dynamics to the hydrodynamic equations by evolving a time-dependent equation of state (EOS) in a manner done previously by Hayes [8]. In this work we build upon the advantages of both of these approaches, having developed a hybrid phase-field/CNT method to describe shock induced phase transitions by coupling the phase field dynamics with the Navier-Stokes hydrodynamic equations. We do this by closely following the work of Anderson *et. al.* [9]. We will present more details of this in a future article.

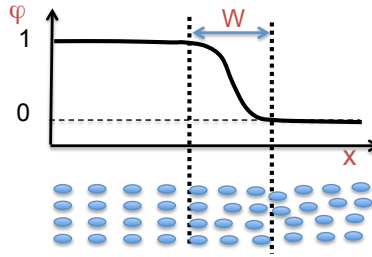


FIGURE 1. A schematic depiction of the atomistic nature of a diffuse solid-liquid interface [2]. The order parameter field ϕ in this case can be thought of as the volume fraction that either phase of the system is occupying at each finite volume element of the system. W is the width of the interface – the “mushy” zone- between the ordered and disordered phase.

Gibbs free energy functional and field evolution equations of a multi-phase system

In the most general case, the global Gibbs free energy for a system with multiple phases is written as [10]:

$$G[\{\phi\}, \nabla\{\phi\}, P, T, \dots] = \int_{\Omega} dV g[\{\phi\}, \nabla\{\phi\}, P, T, \dots], \quad (2)$$

where $\{\phi\}$ is a collection of N phase-field parameters ϕ_i such that $\phi_i \in [0, 1]$ and $\sum_i^N \phi_i = 1$ at a given spatial point and time. In Equation (2), P and T are the pressure and temperature field respectively, whose dynamics are given by hydrodynamic equations. The local Gibbs free energy $g[\{\phi\}, \nabla\{\phi\}, P, T, \dots]$ can be expanded as:

$$g[\{\phi\}, \nabla\{\phi\}, P, T, \dots] = \sum_{i=1}^N g_i^{(1)} + \sum_{i,j(i \neq j)} g_{ij}^{(2)} + \sum_{i,j,k(i \neq j \neq k)} g_{ijk}^{(3)} + \dots, \quad (3)$$

where, $g_i^{(1)}$ is the local Gibbs free-energy of the bulk of each phase i , $g_{ij}^{(2)}$ is the energy penalty associated with the interface between phase i and j , $g_{ijk}^{(3)}$ the energy due to the presence of a three-way contact point between phases i , j and k . Ignoring $g_{ijk}^{(3)}$ and higher order terms simplifies Equations (2) and (3) to:

$$G[\{\phi\}, \nabla\{\phi\}, P, T, \dots] = \int_{\Omega} dV \sum_i^N h_i(\{\phi\}) G_i(P, T) + \sum_{i < j}^N \omega_{ij}(P, T) \phi_i^2 \phi_j^2 + \sum_{i < j}^N \frac{\epsilon_{ij}}{2} \{\phi_i \nabla \phi_j - \phi_j \nabla \phi_i\}^2. \quad (4)$$

The first term on the right hand side of Equation (4) interpolates between the Gibbs free energy $G_i(P, T)$ of each phase i , which can be computed from a multiphase equation of state (EOS) model. The form of the interpolation function

$h_i(\{\phi\})$ is phenomenological. For the simplest case of only two coexisting phases, the most appropriate form is the one due to Langer [11]:

$$h(\phi) = \phi^3 (10 - 15\phi + 6\phi^2). \quad (5)$$

The last two terms on the right hand side of Equation (4) give the explicit expression for $g_{ij}^{(2)}$ in Equation (3). The second term is proportional to the energy barrier, $\omega_{ij}(P, T)$, between bulk phases i and j .

The dynamics of each of these fields is derived by the Euler-Lagrange transport equations in a linear response approach given by:

$$\frac{\partial \phi_i}{\partial t} = -\beta \left[\gamma_i \frac{\delta G[\{\phi\}, \nabla\{\phi\}, P, T, \dots]}{\delta \phi_i} + \xi_i(\{\phi\}, T, P) \right], \quad (6)$$

where γ_i represents the time-scale for atom rearrangements (attachment/detachment) to phase i from all other phases. In Equation (6), $\xi_i(\{\phi\}, T, P)$ represents a stochastic noise term which effectively describes fluctuations that are microscopic in origin. At the coarse-grained scale typical of hydrodynamics present in dynamic experiments (on the order of a micron), we can average over this noise source and utilize classical nucleation theory to provide this mean field. In this context, parameters in our kinetics model determine the surface free energy difference between phases, barrier heights and chemical mobility (which, for solid-solid transformations, has a dependence upon the local sound speed of the initial phase). Coupling the resulting dynamics from Equation (6) to the hydrodynamic equations for P and T , (such as Navier-Stokes) provides one of many possible ways to describe shock-induced phase transitions.

Results from the SAMSA hybrid multi-phase field code

Given its importance for basic planetary science, shock wave propagation has been experimentally studied in a number of settings. While a full introduction to the large data set of dynamically driven Fe experiments is not possible in this letter, we have selected two specific α - ϵ Fe gas-gun impact experiments for which to apply the SAMSA kinetics code: experiments of Jensen *et. al.*[12], and the seminal impact study of Barker and Hollenbach[13]. The experiments of Barker highlight how the shock-wave splits into a leading plastic wave (denoted P1) followed by the phase transition front (P2); further, a third wave arrives due to interaction of a rarefaction from the free surface with the phase interface (PIR, or phase interface reflection wave). Jensen and co-authors undertook shock experiments in which an Fe bullet is fired at a transparent sapphire window in order to directly interrogate the interface that is being driven into the high-pressure ϵ -Fe phase; the results of these experiments show an “overshoot” due to the (it is believed) finite timescale associated with nucleating the new phase, followed by a rarefaction shock due to reversion back into the α -Fe phase.

We explored empirical fitting of the SAMSA kinetics model for all three classes of Fe impact experiment and have found that a single set of parameters can simulate all of these experiments. In order to tune the nucleation rate, we first matched the overshoot profile from the front-surface impact experiments. Next, we tuned the chemical mobility to obtain the best possible match to the Jensen transmission data; the resulting fit is not perfect (especially in the region of the elastic-plastic release) which is not unexpected given that our kinetics model does not yet account for coupling to plasticity fields. Finally, the resulting model matched well the Barker data without further tuning. We also point out that in the case of the Barker data, Boettger and Wallace [5] demonstrated that the effects of phase transition kinetics on the rise-time of the P2 wave is essential to match the PIR, an effect that we discern with SAMSA as well. We have also discovered, however, that the Barker data does not greatly constrain the kinetics parameters, whereas the front surface impact geometry is more amenable to the decoupling of metastability from growth rate.

Summary

We have incorporated a time-dependent Ginzburg-Landau framework to describe shock-induced phase transitions and have implemented that within a new LLNL code named SAMSA. We have used SAMSA to simulate the α - ϵ -Fe non-equilibrium phase transition under three different gas-gun impact conditions and have found good agreement with a single (empirically-fit) model. The essential parameters we varied are the mobility, the nucleation rate and the free energy barrier. Current efforts are focused on extending the Fe model to include very high compression-rate Fe laser experiments, and incorporating atomistic information from molecular dynamics and higher-order terms in the hydrodynamic scale kinetics model.

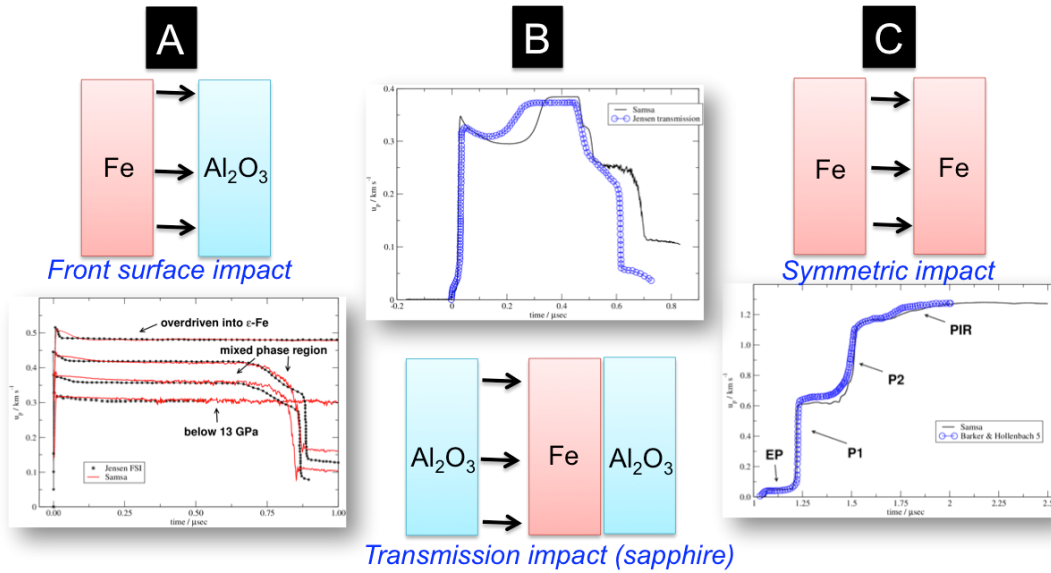


FIGURE 2. Comparisons of simulated velocimetry with SAMSA (black) for three distinct impact experiments of Fe: (a) front surface impact experiments of Jensen[12] where an Fe impactor is launched into a sapphire anvil, (b) wave transmission experiment of Jensen where a sapphire impactor generates a shock in Fe which is tamped with sapphire and (c) symmetric impact experiment of Barker[13] where an Fe impactor generates a shock in Fe and observed at the free surface. Having used the front-surface impact measurement to adjust the nucleation rate, the Jensen transmission experiment was used to adjust the chemical mobility (by matching the rise of the P2 wave) – final application of the resulting model to the symmetric impact case resulted in good agreement without further adjustment.

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